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14. ABSTRACT The overarching goals of the research project include advancing our understanding of strong correlation effects in Mott semiconductors and their response to electric fields. This is relevant to their eventual use as advanced semiconductors in microwave devices or high speed transistors where collective effects are exploited to design switches. The report presents our progress in studying electron transport mechanisms in doped SmNiO <sub>3</sub> . Upon electron doping via hydrogenation, a strongly correlated Mott insulating state is formed in the nickelate. It is therefore important to understand the carrier transport mechanism in the doped nickelate where carriers are strongly					
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## Report Title

Final Report: Research Area 4: Electronics: Mott Transistor: Fundamental Studies and Device Operation Mechanisms

### ABSTRACT

The overarching goals of the research project include advancing our understanding of strong correlation effects in Mott semiconductors and their response to electric fields. This is relevant to their eventual use as advanced semiconductors in microwave devices or high speed transistors where collective effects are exploited to design switches. The report presents our progress in studying electron transport mechanisms in doped SmNiO<sub>3</sub>. Upon electron doping via hydrogenation, a strongly correlated Mott insulating state is formed in the nickelate. It is therefore important to understand the carrier transport mechanism in the doped nickelate where carriers are strongly localized. We present a detailed study of carrier transport as a function of temperature along with description of the mechanisms in a strongly localized picture characteristic of Mott insulators.

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**Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:**

**(a) Papers published in peer-reviewed journals (N/A for none)**

<u>Received</u>	<u>Paper</u>
03/21/2016	2.00 Chen Jikun et al. Self-limited kinetics of electron doping in correlated oxides, Applied Physics Letters, (07 2015): 0. doi: 10.1063/1.4927322
03/21/2016	3.00 Sébastien Cueff, Dongfang Li, You Zhou, Franklin J. Wong, Jonathan A. Kurvits, Shriram Ramanathan, Rashid Zia. Dynamic control of light emission faster than the lifetime limit using VO <sub>2</sub> phase-change, Nature Communications, (10 2015): 0. doi: 10.1038/ncomms9636
03/21/2016	4.00 Jura Rensberg, Shuyan Zhang, You Zhou, Alexander S. McLeod, Christian Schwarz, Michael Goldflam, Mengkun Liu, Jochen Kerbusch, Ronny Nawrodt, Shriram Ramanathan, D. N. Basov, Federico Capasso, Carsten Ronning, Mikhail A. Kats. Active Optical Metasurfaces Based on Defect-Engineered Phase-Transition Materials, Nano Letters, (02 2016): 0. doi: 10.1021/acs.nanolett.5b04122
<b>TOTAL:</b>	<b>3</b>

**Number of Papers published in peer-reviewed journals:**

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**(b) Papers published in non-peer-reviewed journals (N/A for none)**

<u>Received</u>	<u>Paper</u>
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**TOTAL:**

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Number of Presentations: 0.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received      Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received      Paper

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Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received      Paper

08/31/2015    1.00    J. Chen, Y. Zhou, S. Middey, J. Jiang, N. Chen, L. Chen, X. Shi, M. Dobeli, J. Shi, J. Chakhalian, S. Ramanathan. Self-limited kinetics of electron doping in correlated oxides, Applied Physics Letters (07 2015)

TOTAL:      1

Number of Manuscripts:

Books

Received      Book

TOTAL:

Received      Book Chapter

TOTAL:

Patents Submitted

Patents Awarded

Awards

National Academy of Sciences Keck Futures Award, 2014  
Invited Panelist, NSF-SRC Workshop on Energy-Efficient Electronics, DC, 2015

Keynote Speaker, Functional Oxide Thin Films Conference, Mexico (2016)  
Keynote Speaker, Workshop on Electron Correlations, Italy (2015)  
Keynote Presentation, SPIE Defense and Security Conference, Maryland (2014)

Graduate Students

NAME                      PERCENT SUPPORTED

FTE Equivalent:  
Total Number:

### Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Koushik Ramadoss	1.00
<b>FTE Equivalent:</b>	<b>1.00</b>
<b>Total Number:</b>	<b>1</b>

### Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Shriram Ramanathan	0.50	No
<b>FTE Equivalent:</b>	<b>0.50</b>	
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<u>NAME</u>	<u>PERCENT SUPPORTED</u>
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<b>Total Number:</b>	

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### Names of Personnel receiving masters degrees

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### Names of personnel receiving PHDs

<u>NAME</u>
<b>Total Number:</b>

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### Names of other research staff

NAME

PERCENT SUPPORTED

**FTE Equivalent:**

**Total Number:**

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### Sub Contractors (DD882)

### Inventions (DD882)

### Scientific Progress

A technical report is attached to this report submission.

### Technology Transfer

Interaction on basic research topics in Mott semiconductors with industry such as BAE Systems and Micron Technologies.

# **Project Title: Electronics: Mott Transistor: Fundamental studies and device operation mechanisms**

PI: Shriram Ramanathan, Harvard University

Grant Number: W911NF-14-1-0669

## **OVERVIEW**

The overarching goals of the research project include advancing our understanding of strong correlation effects in Mott semiconductors and their response to electric fields. This is relevant to their eventual use as advanced semiconductors in microwave devices or high speed transistors where collective effects are exploited to design switches. The report presents our progress in studying electron transport mechanisms in doped  $\text{SmNiO}_3$ . Upon electron doping via hydrogenation, a strongly correlated Mott insulating state is formed in the nickelate. Since the dopant is in ionic form, an electric field can be utilized to actuate a phase transition independent of temperature. This enables a new form of transistor device. It is therefore important to understand the carrier transport mechanism in the doped nickelate where carriers are strongly localized. We present a detailed study of carrier transport as a function of temperature along with description of the mechanisms in a strongly localized picture characteristic of Mott insulators.

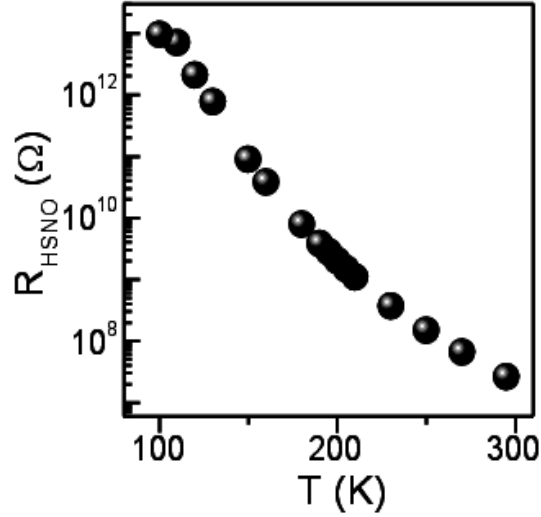
## **I. EXPERIMENTS**

The  $\text{SmNiO}_3$  (SNO) samples were synthesized by co-sputtering from Sm and Ni targets in  $\text{Ar/O}_2$  atmosphere onto a single crystal lanthanum aluminate as substrate [1]. The samples were then lithographically patterned with Pt electrode arrays with width  $\sim 250 \mu\text{m}$  and spacing  $\sim 500 \mu\text{m}$ . The samples were then annealed at  $200^\circ \text{C}$  in hydrogen atmosphere (5%  $\text{H}_2$  in Ar). During this process electron doping occurs via dopant incorporation into SNO lattice [2]. The hydrogenated SNO (H-SNO) sample was then loaded into a probe station for electrical measurements. The current voltage characteristics ( $I - V$ ) were measured at several temperatures between 77 K and 300 K. The sample resistance was determined from the zero bias slope of the  $I - V$  curves.

## **II. RESULTS AND DISCUSSION**

The resistance of H-SNO film as a function of temperature is shown in Fig. 1.

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**Figure 1:** Temperature dependence of resistance of H-SNO showing an insulating behavior

The resistance increases by six orders of magnitude as the sample is cooled down from 300 K to 77 K and shows an insulating behavior. This increase in resistance with reduction in temperature can be viewed in the context of doped semiconductors and thus a possibility of electron hopping between localized states arises. Let us briefly look into various mechanisms leading to hopping transport. It is well known that localized states exist in the forbidden gap of doped semiconductors/disordered insulators. For a semiconductor,  $\sigma \rightarrow 0$  as  $T \rightarrow 0$  K. But at finite temperatures, electrical transport occurs by hopping of charge carriers between nearby localized states and this mechanism is known as nearest neighbor hopping. In such cases, the resistivity ( $\rho(T)$ ) exhibits an activated behavior given by

$$\rho(T) = \rho_0 \exp \left( \frac{\Delta E}{k_B T} \right) \quad (1)$$

where  $\rho_0$  is the prefactor and  $\Delta E$  is the activation energy for hopping. However at low temperatures, it is energetically favorable for an electron to hop to a site that is closer in energy than the nearest neighbor leading to a mechanism known as variable range hopping (VRH) and is given by

$$\rho(T) = \rho_1 \exp \left( \frac{T_0}{T} \right)^p \quad (2)$$

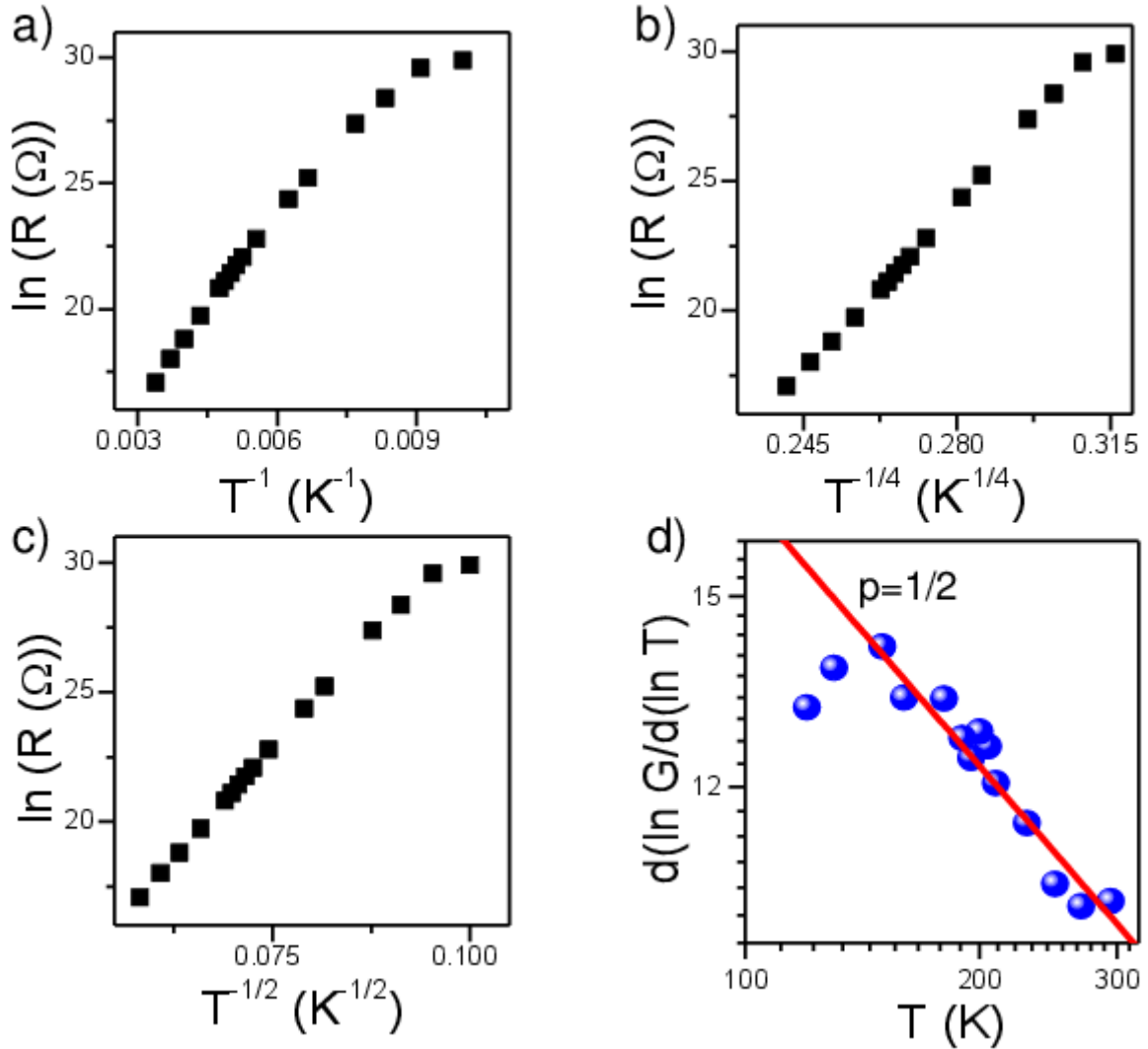
where  $\rho_1$  is the prefactor,  $T_0$  is the characteristic temperature and  $p$  is the exponent dependent on the conduction mechanism. The type of VRH conduction is dependent on the details of the density of states (DOS) around Fermi energy ( $E_F$ ). It was shown by Mott [3] that for a constant DOS,  $p = 1/(D + 1)$  where  $D$  is the dimensionality of the system. In three dimensions,  $p = 1/4$  and  $T_0$  is given by [4, 5]

$$T_0 \equiv T_M = \frac{18}{k_B N(E_F) \xi^3} \quad (3)$$

where  $N(E_F)$  is the DOS near  $E_F$  and  $\xi$  is the localization length. But when Coulomb interaction between charge carriers is taken into account, a gap appears in the DOS near  $E_F$  and one can show that  $p = 1/2$ . This mechanism is known as Efros-Shklovskii (ES) VRH in which  $T_0$  is given as [6]

$$T_0 \equiv T_{ES} = \frac{2.8 e^2}{4\pi\epsilon k_B \xi} \quad (4)$$

where  $\epsilon$  is the dielectric permittivity.



**Figure 2:** a) Plot of  $\ln(R)$  vs  $1/T$  for H-SNO film to check for nearest neighbor hopping transport mechanism. b) Plot of  $\ln(R)$  vs  $T^{1/4}$  for H-SNO film to check for Mott VRH. c) Plot of  $\ln(R)$  vs  $T^{1/2}$  for H-SNO film to check for ES VRH. d) RCDA

of conductance shows a slope of  $p = 1/2$  indicative of ES VRH mechanism for H-SNO.

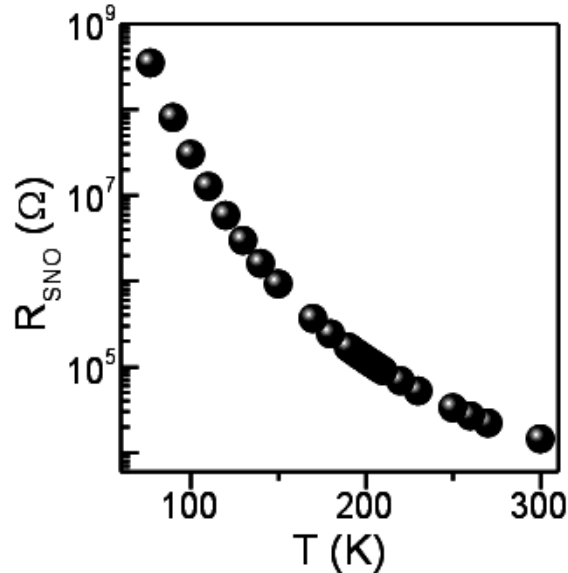
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For H-SNO samples, the resistance data shows a clear deviation from activated transport mechanism (Fig. 2a). The data shows a linear behavior for both Mott and ES VRH mechanisms (Fig. 2b,c). In such cases, the exponent  $p$  can be determined unambiguously by a method based on resistance curve derivative analysis (RCDA) [7]. The essence of this method is to calculate a parameter  $w(T) = \frac{d \ln G}{d \ln T}$ , where the conductance  $G = 1/R$ . After some algebra, it can be shown that

$$w = p \left( \frac{T_0}{T} \right)^p \quad (5)$$

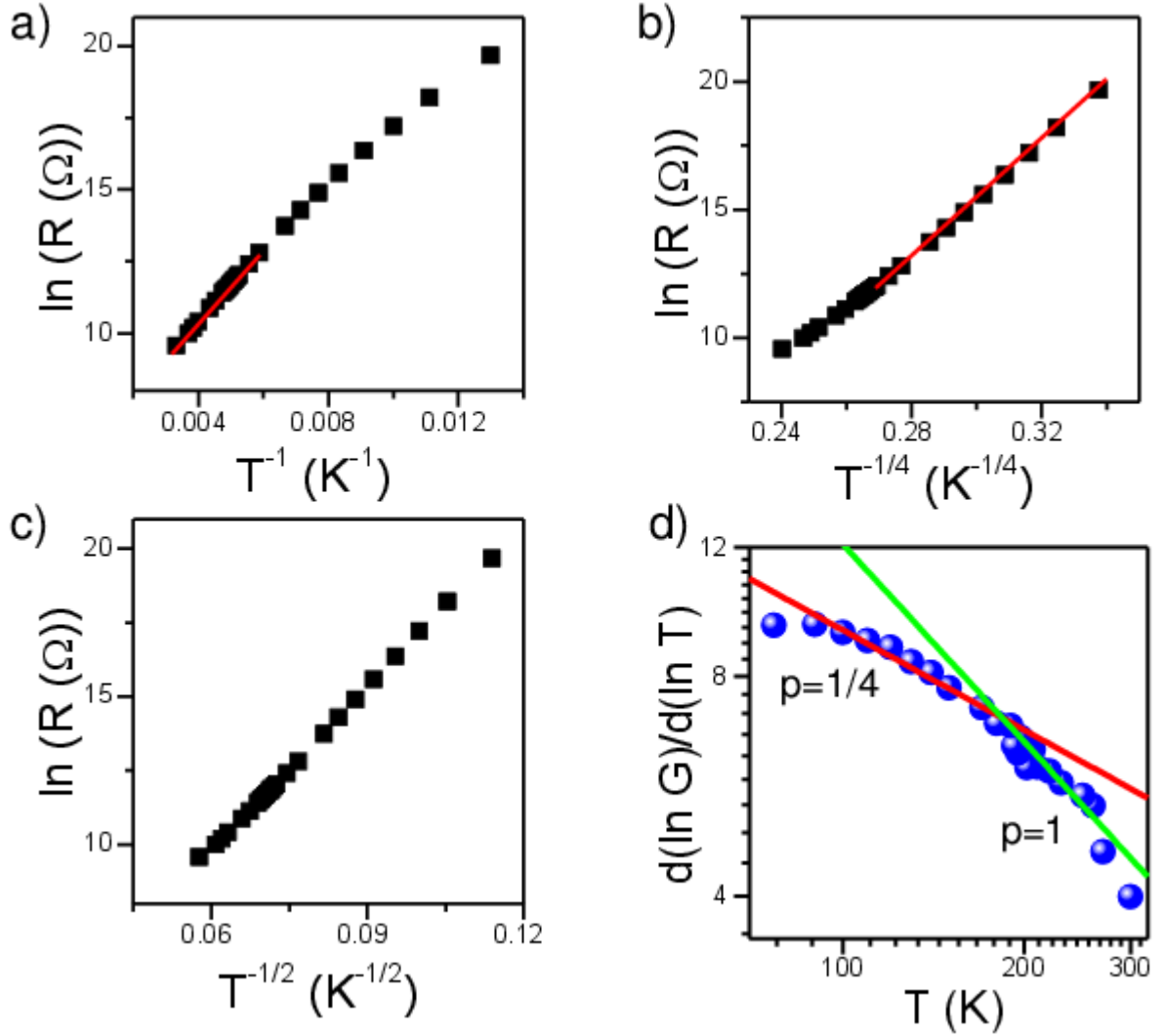
A log-log plot of Eqn. 5 yields the exponent  $p$  which is determined from the slope. Various transport mechanisms like NNH, ES VRH and Mott VRH correspond to value of  $p = 1$ ,  $1/2$  and  $1/4$  respectively. A plot of  $w$  is shown in Fig. 2d from which the slope is determined to be  $1/2$ . Thus the dominant mechanism in H-SNO over the entire temperature range is ES VRH signifying the presence of Coulomb interactions in the system. From a linear fit to Fig. 2c yields  $T_{ES} = 1.194 \times 10^5$  K and  $R_v = 49.3$  m $\Omega$ . Using Eqn. 4 and taking the dielectric constant of H-SNO to be  $\sim 5$  [2], we determine  $\xi = 0.78$  Å which is much smaller compared to the lattice spacing of bulk SNO (0.3796 nm). Thus the carriers are strongly localized within a unit cell.

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**Figure 3:** Temperature dependence of resistance of SNO showing an insulating behavior

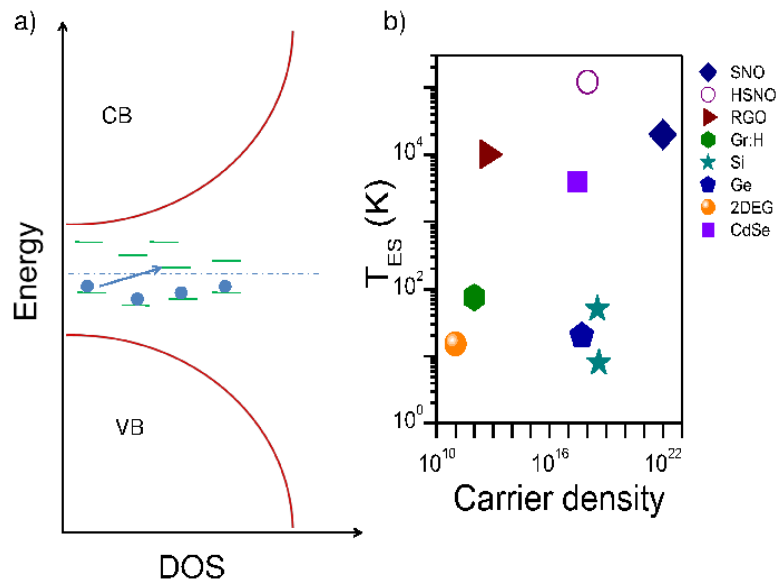
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**Figure 4:** a) Plot of  $\ln(R)$  vs  $1/T$  for SNO film to check for nearest neighbor hopping transport mechanism. b) Plot of  $\ln(R)$  vs  $T^{-1/4}$  for SNO film to check for Mott VRH. c) Plot of  $\ln(R)$  vs  $T^{-1/2}$  for SNO film to check for ES VRH. d) RCDA of conductance of SNO film shows a slope of  $p = 1/4$  and 1 indicative of Mott VRH and activated transport mechanism respectively.

In order to develop a better understanding of the transport properties of H-SNO, we performed electrical measurements on a SNO sample which was obtained by annealing H-SNO to 573 K for 2 hours in air. Subsequently the sample resistance was measured as shown in Fig. 3. It is important to note that the resistance of SNO at 300 K drops to  $\sim 10$  K $\Omega$  compared to  $\sim 20$  M $\Omega$  for H-SNO. The sample exhibits 5 orders of magnitude change in resistance over the measurement temperature range. The plots of resistance for possible mechanisms like NNH and VRH are shown in Fig. 4a-c. We use RCDA to show that the transport is governed by activated type behavior ( $p = 1$  in Fig. 4d) for  $T \gtrsim 180$  K with an activation energy  $\sim 110$  meV and Mott VRH ( $p = 1/4$  in Fig. 4d) at low temperatures. We find the characteristic temperature  $T_M = 1.74 \times 10^8$  K which yields  $N(E_F) = 1.2 \times 10^{18}$  eV<sup>-1</sup>cm<sup>-3</sup>. Thus we find that the transport mechanism is distinct for H-SNO in comparison to pristine samples. The transport mechanism in SNO switches from NNH to Mott VRH as the

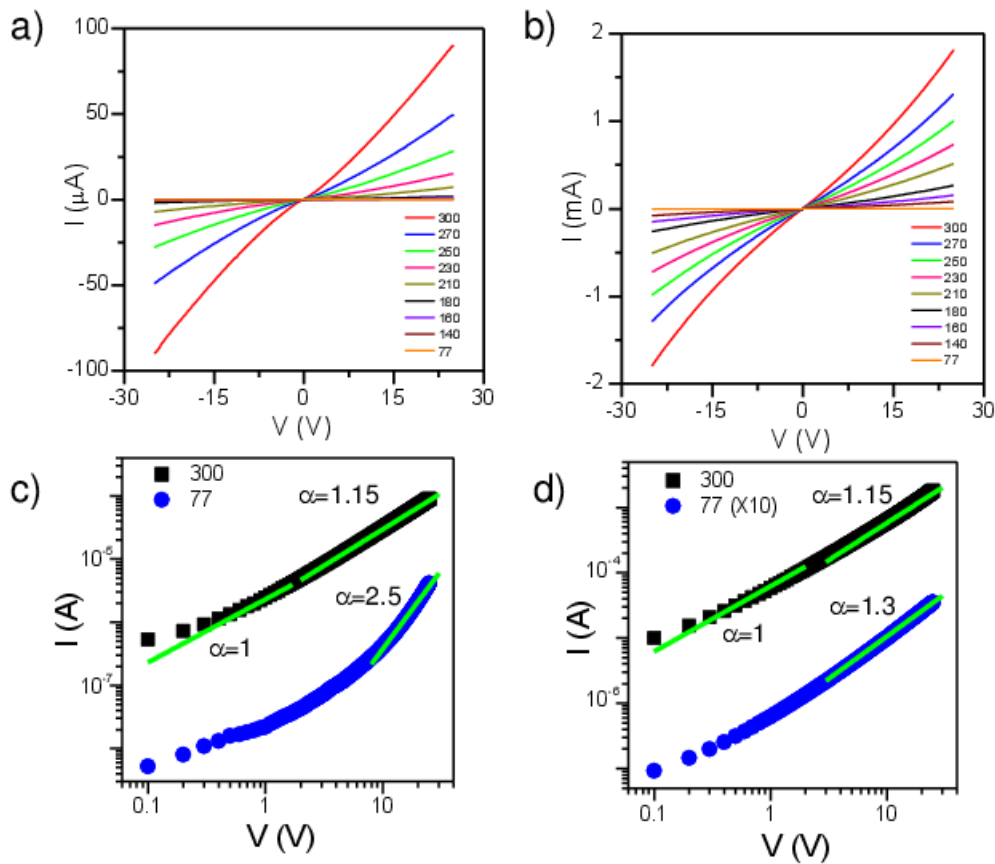
temperature is lowered similar to other crystalline and amorphous semiconductors [8–10]. However hydrogen incorporation into SNO has a distinct effect unlike in conventional semiconducting systems where hydrogen doping introduces compensation decreasing the carrier density. In H-SNO, hydrogen loses an electron which changes the valency state of the  $\text{Ni}^{3+}$  to  $\text{Ni}^{2+}$ . The proton ( $\text{H}^+$ ) then goes into lattice preferentially closer towards the ligands serving as a source of compensation. Earlier studies on H-SNO indicate that the hydrogenation process increases the bandgap of the system [2]. Thus one can picture the Fermi energy to be located deep in the forbidden band amidst the localized states (Fig. 5a) leading to a large increase in resistance. The  $\text{H}^+$  ions increase the disorder and thus correlation effects become important thereby leading to ES VRH as a dominant mechanism of carrier transport.



**Figure 5:** a) Schematic representation of band diagram (energy vs DOS) of a disordered semiconductor with Fermi energy (dashed blue line). Green lines indicate impurity states in the bandgap and blue circles represent electrons. The hopping of electrons between impurity states is represented by a blue arrow. b) Comparison of  $T_{ES}$  for various semiconductors H-SNO (this work indicated by open circle), SNO [11], Reduced graphene oxide (RGO) [12], hydrogenated graphene (Gr:H) [13], Si [14], Ge [15], GaAs/AlGaAs heterostructure (2DEG) [16], CdSe [17]. Carrier density in units of  $\text{cm}^{-3}$  except for 2DEG, RGO and Gr:H (units of  $\text{cm}^{-2}$ ).

In Fig. 5b, we compare  $T_{ES}$  of H-SNO with that of other semiconductors from literature. We find that for similar carrier densities, H-SNO exhibits a larger  $T_{ES}$  compared to other systems. This is clear from the variation of resistance (five orders change) over the entire temperature range (77 - 300 K). A large  $T_{ES}$  is directly correlated with the strong localization of carriers (Eqn. 4). The number density for H-SNO is estimated from a simple Drude model of conduction by comparing it with pristine sample. It is known that for a-Si films [18], the Coulomb interaction is stronger when the charge carriers are localized in band-tail states which makes the distribution of these states to lie deep in the bandgap of the

material. One can think of a similar scenario in the case of H-SNO which has a larger bandgap ( $\sim 3$  eV) and carriers are strongly localized.

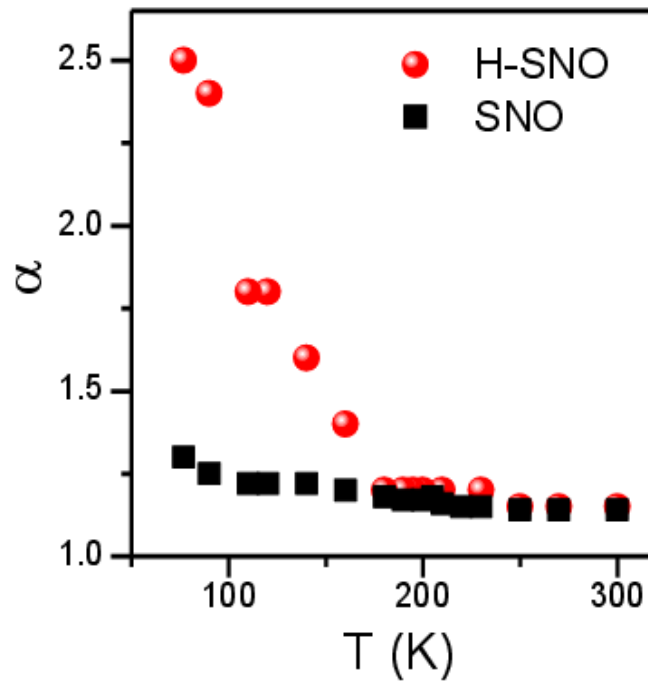


**Figure 6:** a)  $I$  -  $V$  characteristics of H-SNO film at various temperatures. The curves become non-linear for  $V \gtrsim 5$  V. b)  $I$  -  $V$  characteristics of SNO film at various temperatures. The curves become non-linear for  $V \gtrsim 5$  V. c) Log-log plot  $I$  -  $V$  characteristics of H-SNO film for two different temperatures (77 K and 300 K). The exponent  $\alpha$  is also indicated for both low and high fields. d) Log-log plot  $I$  -  $V$  characteristics of SNO film for two different temperatures (77 K and 300 K). The exponent  $\alpha$  is also indicated for both low and high field.

Finally we compare the  $I$ - $V$  curves of SNO and H-SNO at high fields ( $V \gtrsim 5$  V). As shown in Fig. 6a,b, both H-SNO and SNO display non linear  $I$  -  $V$  characteristics over entire temperature range. We find that the current is linear in voltage for  $V \lesssim 5$  V and exhibits  $I \propto V^\alpha$  behavior at larger voltages (Fig. 6c,d). The exponent  $\alpha$  increases with decrease in temperature for both H-SNO. In Fig. 7,  $\alpha$  is shown as a function of temperature for both the samples. We find that  $\alpha$  displays a larger variation from 1.1 to 2.5 for H-SNO samples which are strongly insulating whereas in the case of SNO  $\alpha$  shows a much smaller variation ( $\sim (1.1 - 1.3)$ ). Any attempts to fit our data with conduction mechanisms like Schottky emission, Fowler-Nordheim tunneling, Poole-Frenkel emission failed leaving us with a possibility of space-charge limited conduction (SCLC). Previous experiments on epitaxial SNO thin films [19] have shown power law behavior in  $I$  -  $V$  characteristics ( $\alpha \sim 8$ ) across

the metal to insulator regime of SNO and the results were explained in terms of SCLC caused due to trap states located in the bandgap. Thus one can interpret the large increase in  $\alpha$  in H-SNO at low temperatures as being caused due to the dominance of trap states which localize free carriers thereby enter SCLC regime with  $\alpha > 2$ .

To summarize, we have presented the transport measurements on H-SNO where the conduction mechanism is dominated by an interaction mediated hopping of charge carriers which are strongly localized in mid gap states.



**Figure 7:** Comparison of  $\alpha$  for H-SNO and SNO films over the temperature range 77 - 300 K.

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